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Gas: A Neglected Phase in Remediation of Metals and Radionuclides

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“It is our business to puncture gas bags and discover the seeds of truth.” – Virginia Woolf

The gas phase is generally ignored in remediation of metals and radionuclides because it is assumed that there is no efficient way to exploit it. In the literal sense, all remediations involve the gas phase because this phase is linked to the liquid and solid phases by vapor pressure and thermodynamic relationships. Remediation methods that specifically use the gas phase as a central feature have primarily targeted volatile organic contaminants, not metals and radionuclides. Unlike many organic contaminants, the vapor pressure and Henry’s Law constants of metals and radionuclides are not generally conducive to direct air stripping of dissolved contaminants. Nevertheless, the gas phase can play an important role in remediation of inorganic contaminants and provide opportunities for efficient, cost effective remediation. Our objective here is to explore ways in which manipulation of the gas phase can be used to facilitate remediation of metals and radionuclides.

Components in the gas phase are linked to their counterparts in the aqueous phase by a partitioning relationship such as Henry’s Law:

$$P = HC_w$$

where P is the partial pressure of the component in the gas phase, H is the Henry’s Law constant for the component, and C_w is the concentration of the component in the aqueous phase. Typical

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units for Henry's Law constant are $\frac{atm \cdot m^3}{mole}$, though other units are used where convenient.

Volatility from water has been divided by Lyman et al. (1) into high ($H > 10^{-3}$), medium ($3 \times 10^{-7} < H < 10^{-3}$), and low ($H < 3 \times 10^{-7}$). Behavior of a gas in water must be considered when assessing volatility. Some gases react in water (e.g., CO_2 , H_2S , O_3 , I_2 , NH_3) and others remain structurally unchanged (e.g., O_2 , H_2 , N_2 , CH_4). If a gas reacts in water, then further dissolved species are generated that influence the overall mass transfer. These interactions in the aqueous phase are assumed to be in accordance with thermodynamic and kinetic controls and are predictable.

Discussion of remediation involving a gas phase highlights an important difference between most common organic contaminants and most inorganic contaminants. That is, the chemical properties of dissolved inorganic contaminants are much more likely than organic contaminants to be affected by solution chemistry – sometimes providing opportunities and sometimes challenges. As in any remediation, it is important to understand solution chemistry and its effect on the speciation of the contaminant of interest. These various factors, all of which are studied and documented in the literature, control the potential uses of the gas phase for environmental remediation of metals and radionuclides.

Figure 1 divides the use of the gas phase in remediation into three broad classes: 1) Direct removal of contaminant vapor from groundwater, 2) Manipulation of master geochemical variables, and 3) Injection of gaseous stabilizing agents. All three involve injection and/or extraction of a gas phase. Systems may use a standard gas such as air nitrogen, a specialty mixture of gases, or liquid reagents in combination with gas injection/extraction.

The gas phase can be used in a typical *ex situ* treatment plant or *in situ*, in the environment.

When used *ex situ*, gas phase concepts can be designed and implemented using standard chemical and environmental engineering principles. When used *in situ*, gas phase concepts have many advantages. Injection of gases is simple and inexpensive and can be performed in the vadose zone, the groundwater, and in aquatic systems. *In situ* treatment methods require controlled uniform delivery of amendments throughout the contaminated zone. Gas can more effectively penetrate low permeability materials and tends to cover a larger area in many settings. Notably, the injection of gas in the vadose zone, a common and significant environment for residual metal and radionuclide contamination, is more efficient and predictable than injection of liquid or solid reagents. The potential value of gas phase removal or stabilization methods for metals and radionuclide contaminants is presaged in the past success of bioventing and biosparging, two common and widely used bioremediation methods for organic contaminants. These cost-effective methods rely on the engineered movement of air through the vadose zone and shallow groundwater to deliver oxygen and accelerate degradation of organic contaminants.

Direct Removal of Contaminant Vapor

The simplest and most effective treatment for metals and radionuclides would be removal of the contaminant from the subsurface by vapor extraction. The major advantage of this is that the contaminant is actually removed from the subsurface system, rather than merely stabilized in place. This approach requires that the contaminant be in a volatile form that is present in the vapor phase in the vadose zone or, if the contaminant is dissolved, has a Henry's Law constant that is conducive to

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stripping. Unfortunately, few metals and radionuclides have appropriate chemical and physical properties for use of vapor extraction.

The Henry's Law constant is a primary controlling factor in the mass transfer of a volatile contaminant from solution to the vapor phase. For efficient remediation by air stripping, contaminants should have a Henry's Law constant greater than about 10^{-4} atm-m³/mole.

Reasonable estimates of Henry's Law constants can be made from the ratio of a constituent's vapor pressure to its solubility (1, 2). Figure 2 shows the vapor pressures for several constituents plotted versus their solubilities, with the Henry's Law constants represented by the diagonal lines. The volatile organic contaminants carbon tetrachloride (CCl₄), trichloroethylene (TCE), and perchloroethylene (PCE) are commonly air stripped from groundwater and are shown for comparison with elemental mercury (Hg⁰), elemental iodine-129 (¹²⁹I₂⁰), and carbon-14 (as ¹⁴CO₂).

The figure illustrates an important point about Henry's Law, that it is not controlled solely by vapor pressure. The Henry's Law constants for Hg⁰ and CO₂ are similar, yet the vapor pressure for Hg⁰ is several orders of magnitude lower than that for CO₂. However, the solubility of Hg⁰ is also several orders of magnitude lower, and thus the ratios – the Henry's Law constants – are similar. The estimated Henry's Law constant for I₂⁰ is about 40 times lower than for Hg⁰, but is still high enough to allow air stripping in some cases.

In general, simple chemical manipulations are required to convert the dominant aqueous forms of these inorganic contaminants into forms that are air strippable. Figure 3 shows the fields of

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dominance for the strippable forms – Hg^0 , I_2^0 and CO_2 – on a pe-pH diagram with the ferric-

ferrous iron couple shown for comparison. The figure illustrates that the strippable forms of all three contaminants exist at conditions that can be created in groundwater with simple single-step chemical manipulations.

Though some mercury is disposed as elemental and can be directly air stripped, in natural systems most exists in groundwater as the oxidized form, Hg(II) . However, dissolved Hg(II) is readily reduced to strippable Hg^0 . Based on analytical methods for mercury analysis (3, 4) and preliminary work by Southworth (5), Looney et al. (6) demonstrated that stannous chloride added to groundwater converts Hg(II) to Hg^0 allowing the mercury to be stripped from solution by air. For groundwater with low levels of mercury contamination, low doses of stannous chloride, near stoichiometry, were effective at reducing Hg(II) to the strippable form allowing low cost air stripping of the contaminant.

Iodine-129 is a product of uranium fission in reactors. When present in the subsurface, it is significant because it has a long half-life (1.6×10^7 years), is mobile in groundwater, and bioconcentrates in the thyroid gland. Virtually all iodine-129 disposed to the subsurface is in the reduced form $^{129}\text{I}^-$, and a complex mixture of related aqueous species. These cannot be directly air stripped. However, under the right conditions, iodine-129 can be oxidized to $^{129}\text{I}_2^0$, a form that is readily strippable. As Figure 3 shows, these conditions are acidic and relatively oxidizing. Interestingly, preliminary studies by the authors demonstrated that sparging with a mixture of air and ozone efficiently removed iodine from water. In contrast, little stripping of I_2 was observed in experiments where hydrogen peroxide and/or sodium persulfate solutions were used as

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oxidants. We speculate that oxidation occurring at the gas-water interface may have some kinetic advantages, allowing rapid mass transfer of I_2^0 from the aqueous phase to the gas phase, preventing competing reactions that form nonvolatile species such as I_3^- which are not strippable. Thus, stripping with an ozone-air mixture is a potentially useful method for removing this difficult contaminant from groundwater.

At some nuclear sites ^{14}C is a concern because of its long half-life (5730 years) and high bioconcentration factor. Depending on pH and redox conditions, ^{14}C generally exists in groundwater as organic matter, light hydrocarbons, or as inorganic species such as dissolved $^{14}CO_2$, bicarbonate, or carbonate. For systems dominated by inorganic carbon, in situ stripping is viable. To efficiently strip ^{14}C from water the dominant species must be dissolved CO_2 , which occurs at pH values less than 6 (Figure 3). If groundwater pH exceeds 6, it can be decreased by injection of air containing a small amount of acid vapor (e.g., HCl), altering the inorganic carbon speciation to favor stripping. Alternatively, at pH values just above 6, CO_2 fortified air could be used to lower the pH of the groundwater and strip the radioactive CO_2 .

Using the gas phase to directly strip mercury, ^{129}I , and ^{14}C represents a promising alternative to traditional remediation methods for these contaminants. All have been remediated using pump-and-treat methods that involve ion exchange, adsorption, or precipitation. The problem with these methods is the disposition of the treated water and the volumes of solid waste created. In situ air stripping shares the advantage of pump-and-treat methods in that it removes the contaminant from the aquifer rather than just stabilizing it in place, but produces no waste water and is expected to produce lower volumes of solid waste. Ion exchange, adsorption, and

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precipitation are, in general, not highly selective and many constituents other than the targeted ones are removed. In contrast, air stripping of these contaminants is highly selective. Thus, even though a canister of solid material may be required at the surface to remove the contaminant from extracted vapor, much less material would be used. This is because the masses of contaminant and other stripped constituents are low compared to the masses of non-target constituents removed from groundwater by an ion exchange system.

Manipulation of Master Variables

Most inorganic contaminants cannot be removed from an aquifer as a vapor, but the transport behavior of many respond to changes in the master variables -- pH and redox potential. Many contaminants are redox sensitive (e.g., uranium, plutonium, technetium-99, chromium, arsenic, and copper) with one valence state being more mobile than the others. Likewise, solubility and adsorption of most inorganic contaminants vary with pH. Both master variables can be manipulated using gas phase reagents to stabilize and sequester the contaminant in place. This general approach is being developed and tested in several laboratories.

Varying the partial pressure of carbon dioxide (PCO_2) in the subsurface is an easy way to manipulate pH. For a contaminant plume with pH between 6 and 10 the pH is generally related to PCO_2 by the equation:

$$pH = -\log\left(\frac{PCO_2}{Alk}\right) + 7.8$$

where *Alk* is the carbonate alkalinity in moles/Liter. This assumes for simplicity that weak acids other than bicarbonate are not present. Most subsurface environments have much higher PCO_2 values than atmospheric air. Thus, injection of air into many plumes will raise the pH enough to

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affect mobility of several contaminants. Figure 4 shows the fraction of strontium adsorbed versus PCO_2 in a plume that is dominated by carbonate species. If the initial PCO_2 is near 0.01 atm, then injecting atmospheric air will increase the fraction of strontium adsorbed to about 45%, path A-A' on Figure 4. This represents a change in K_d from about 0.2 to 14 ml/g. Using the standard retardation equation (7), a bulk soil density of 1.6 g/cm^3 , and an effective porosity of 0.2, this is the equivalent of increasing the retardation factor from 2.6 to 113. This is ideal for strontium-90 because it has a relatively short half-life of 28.6 years. Thus, the increase in travel time caused by lowering the PCO_2 of the aquifer could allow strontium-90 to decay to negligible concentrations before reaching an exposure point.

Solubility of many mineral phases that control contaminant concentrations is also affected by pH, and thus can be manipulated by varying PCO_2 . Figure 4 shows the solubility of the lead carbonate mineral, cerussite, versus PCO_2 . In groundwater at a PCO_2 of 0.01 atm the solubility of cerussite can be reduced from 350 ug/L to 110 ug/L by sparging with untreated air (path B-B' on Figure 4). Many other contaminant stabilizing phases, including oxides and phosphates, are pH sensitive and become less soluble at lower PCO_2 values.

Redox potential can also be manipulated by injection of gaseous reagents. Most redox sensitive metals and radionuclides are less mobile under reducing conditions. For example, the U(VI) phase schoepite ($\text{UO}_3 \cdot 2\text{H}_2\text{O}$) is several orders of magnitude more soluble than the U(IV) phase uraninite (UO_2). Hence, it is beneficial to establish reducing conditions in uranium plumes to minimize mobility. This can be achieved by injecting gases, in particular those that promote anaerobic microbial activity such as combinations of N_2 , H_2 , and CH_4 . H_2 gas has been injected

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in the subsurface to promote reductive dechlorination of solvents (8). If there is a carbon source in the aquifer, using H_2 as an electron donor will stimulate anaerobic bacteria and produce reducing conditions favorable to metal stabilization. H_2 gas can also be introduced to the subsurface without direct injection by deploying hollow-fiber membranes (9).

Establishing oxidizing conditions in an aquifer can also be useful in remediation of metals and radionuclides. Some constituents such as arsenic may be less mobile in the oxidized form. Of more general use is the oxidation of ferrous to ferric iron to force precipitation of ferric oxyhydroxide phases. In acid mine drainage plumes ferrous iron is often present in hundreds of mg/L. Oxidation of the ferrous iron can force precipitation of ferric iron phases that can act as carriers for co-precipitation of some metals and as an adsorbent for others. This may also reduce permeability which can be beneficial at some sites and detrimental at others.

Oxidizing conditions can be created in the subsurface using either gaseous oxygen or ozone. Gaseous oxygen produces a slower response because of its limited solubility and slower reaction kinetics than for the ozone reaction. Ozone has been used extensively for oxidizing organic contaminants, often in combination with other oxidants or radiation, and is well established commercially. Its use at metal and radionuclide sites would be similar with adjustments for amount of ozone required. Ozone must be made on site, but this eliminates the need for transporting potentially dangerous reagents.

A third master variable that can affect metal and radionuclide contaminants in the vadose zone is moisture content. Water vapor can be removed by injection of dry air and extraction of moist air

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beneath an infiltration barrier. The decreased moisture content minimizes migration of metal-bearing water to the water table and causes chemical effects that tend to stabilize metals and radionuclides. Capillary barriers have been discussed previously (e.g., 10) , but these rely on grain-size differences to keep a volume dry rather than injection of air. The chemical effects induced by desiccation are predominantly precipitation or co-precipitation and enhanced adsorption of contaminants. Precipitation occurs because the removal of water increases concentrations of constituents to the point that many exceed their solubility. Adsorption of some metals and radionuclides may increase in response to pH changes caused by the lower PCO_2 in the drying air. Moisture removal, or desiccation, is currently being examined for remediation of the deep vadose zone at the Department of Energy reservation at Hanford Washington .

Manipulation of master variables may or may not permanently stabilize long-lived contaminants. For example, manipulation of pH by varying PCO_2 is a permanent solution for strontium-90 contamination because the increased retardation of strontium-90 and other short-lived radionuclides (e.g., cobalt-60, plutonium 238) may allow them to decay to acceptable concentrations before reaching exposure points. Manipulation of master variables is generally not a permanent solution for stable or long-lived contaminants because stabilized contaminants will begin to remobilize when active manipulation stops. The exceptions are cases where manipulations bring the master variables closer to natural conditions. However, if the manipulation moves the system away from natural conditions, then remobilization is likely when active manipulation ceases and conditions move back toward natural. The question that must be answered for these systems is whether remobilization will occur at a sufficient rate to cause concentrations in groundwater to exceed regulatory standards.

Injection of Gaseous Stabilizing Agents

The third class of remediation methods exploiting the gas phase is injection of gaseous stabilizing agents. The reagents should contain anions that form insoluble salts with the contaminants. Two examples are phosphate and sulfide. Phosphate in various forms has been successful at stabilizing metals such as lead, cadmium, and uranium as insoluble phosphate phases. Likewise, sulfide is an effective stabilizer of metals such as lead, zinc, mercury, copper, and nickel.

Most efforts at stabilization of metals with phosphate have involved solid forms, but phosphate can also be injected as a gaseous phase. The PHOSter® system (11) has been successfully used to stimulate bioremediation of organic contaminants. It provides phosphorus, a limiting nutrient, to the subsurface as a gaseous soluble organic phosphate. Bacteria break down the organic phosphate for its nutrient value, but some phosphate ion is released to groundwater. This is then available to react with metals and radionuclides to form stable insoluble phosphate phases. The contaminant may precipitate by abiotic reaction with dissolved phosphate or may be stabilized within bacterial cells as a phosphate (12, 13, 14). Phosphate released by the biomass may also adsorb to soil and enhance the adsorption of metals and radionuclides (15, 16). This may be an excellent method of stabilizing lead from leaded-gasoline plumes.

Metals and radionuclides have also been stabilized with sulfide, either generated microbially in situ or injected as H₂S gas. Laboratory and field experiments (17, 18) indicate that Cr(VI) and

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U(VI) can be reduced to less mobile forms by injection of H₂S gas into the subsurface. Injection of H₂S could also cause direct precipitation of low solubility sulfides of other metals, though Thornton and Amonette (18) report that copper was the only non-target metal that precipitated in their experiments.

Longevity of immobilization by injection of gaseous stabilizing agents may vary considerably. Injection of hydrogen sulfide into a normally oxidized system is not likely to stabilize contaminants indefinitely without some means of keeping the system reduced. In contrast, injection of hydrogen sulfide into a naturally reduced system may stabilize contaminants for the long-term. Injection of a gaseous phosphate may be longer lived because there will be natural phosphate in the system when it returns to normal conditions. This would slow dissolution of contaminant-phosphate phases. This is evidenced by the longevity of natural phosphate phases such as apatite that persist over geologic time.

To Gas or Not To Gas

Remediation strategies for metal and radionuclide contamination that involve the gas phase are useful for many problems, but are not universally applicable. As with other types of remediation, understanding site specific properties is critical to evaluating use of clean-up methods involving gas. Site geology controls the delivery and distribution of reagents in the subsurface, and at many sites injection of liquids or solid slurries may be as viable as gas injection. Site mineralogy is particularly important in remediations involving pH or redox manipulation, because minerals can buffer pH and redox conditions making manipulation of these variables difficult. Likewise, groundwater composition is critical because it controls contaminant speciation, and thus response

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to treatment. Figure 5 summarizes various in situ remediation strategies discussed, their potential target contaminants, the site characteristics that would limit use of the method, and their stage of development. A limiting factor for all is access, which includes both interfering man-made structures and limiting geological features such as low permeability. Access is critical because all of the remediation technologies require that mass be either injected into or removed from the contaminated zone. There are other remediation strategies involving the gas phase, but it is hoped that the discussion presented here will serve as a reminder that exploitation of the gas phase in remediation of metal and radionuclide contamination is worth consideration.

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3. Fields of dominance on a pe-pH plot for air strippable forms of mercury, iodine-129, and carbon-14.
4. Calculated adsorption of strontium on hydrous ferric oxide and solubility of mineral cerussite (PbCO_3) as a function of PCO_2 .
5. Summary of gas phase remediation methods.

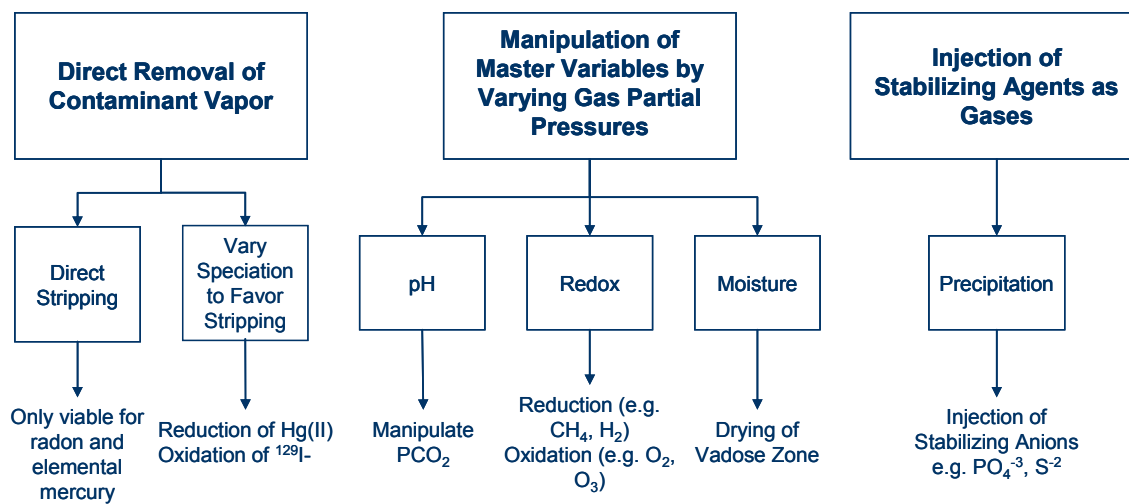


Figure 1

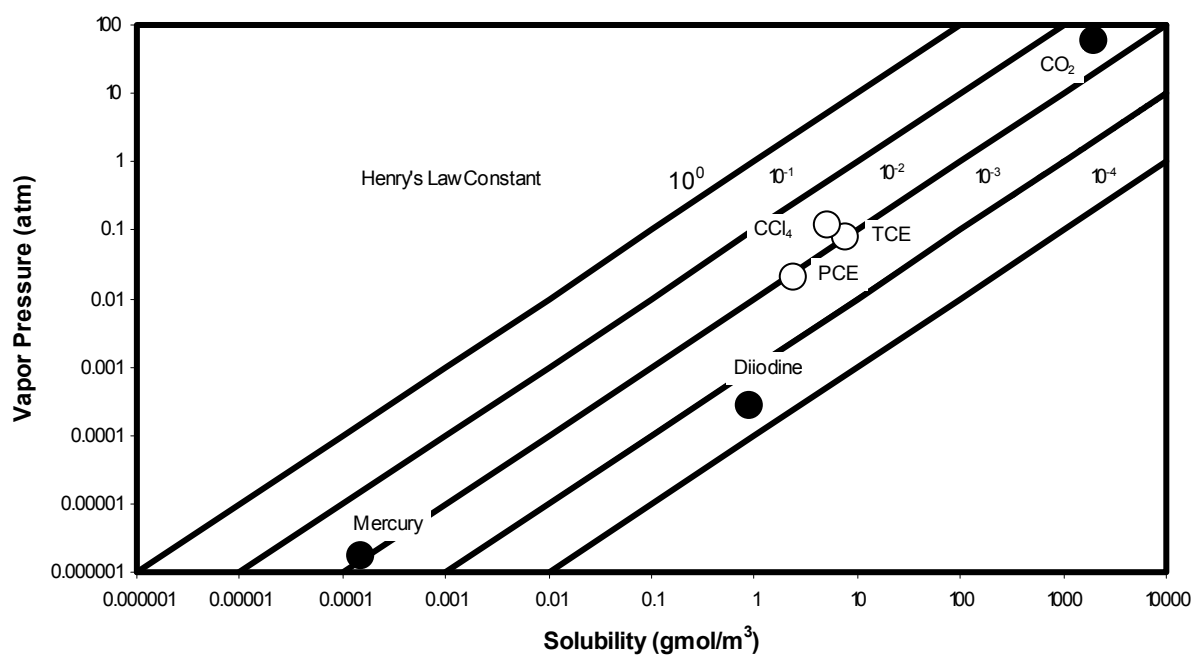


Figure 2

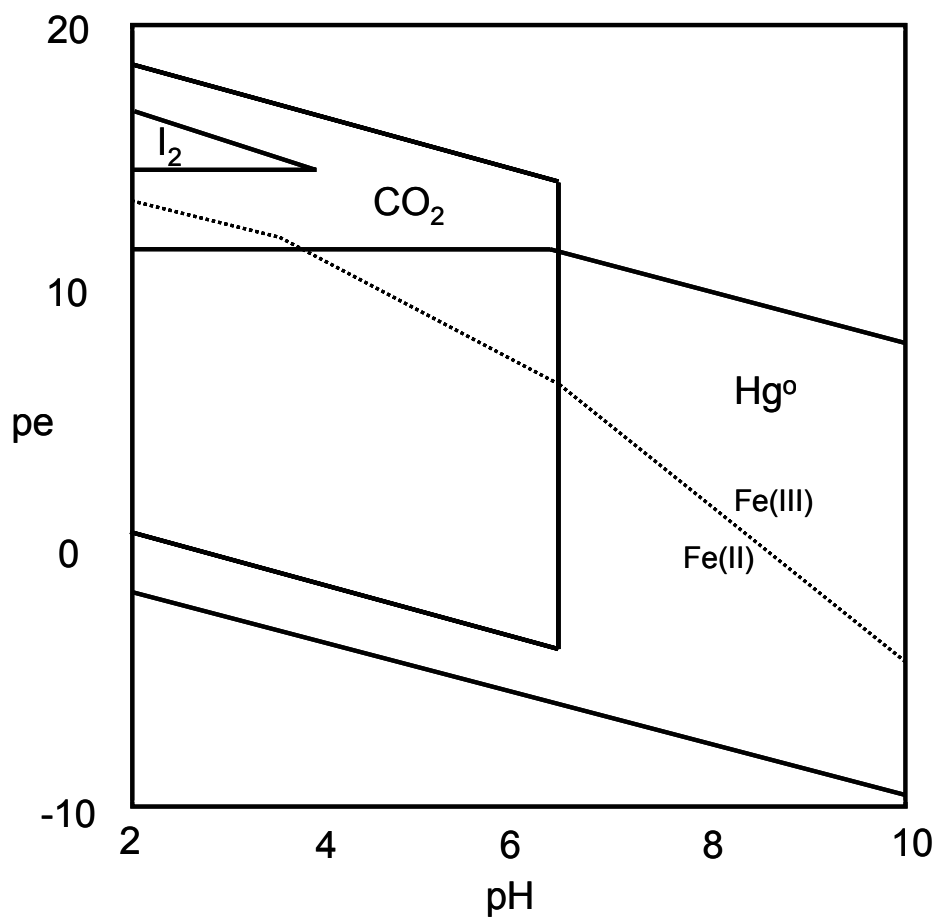


Figure 3

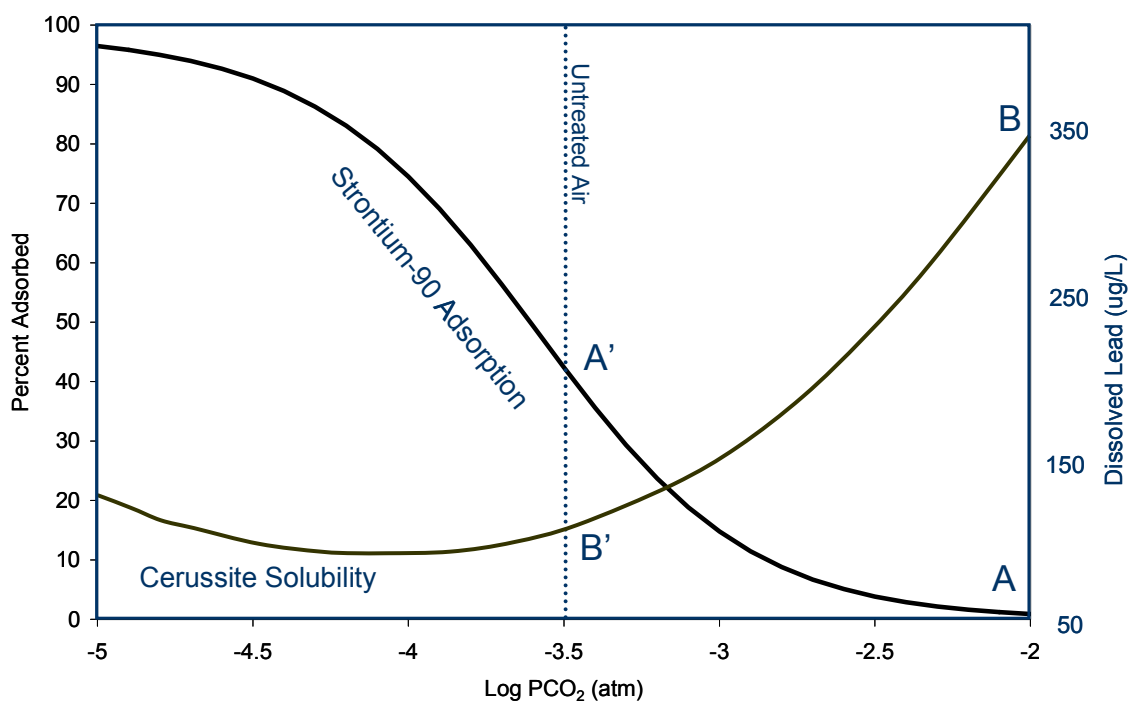


Figure 4

Category	Specific Method	Potential Target Contaminants	Primary Limiting Factors (<i>in situ</i>)	Development Stage
Direct Removal	Chemical Manipulation + Air Sparging	Hg, ¹²⁹ I, ¹⁴ C, ^{220/222} Rn	plume composition,	Lab tested, ready for <i>ex situ</i> use for some contaminants
Master Variable Manipulation	pH Adjustment	Many	plume composition, mineralogy	Ready for field testing
Master Variable Manipulation	Redox Adjustment	U, Cr, As, ⁹⁹ Tc	mineralogy	Commercially available
Master Variable Manipulation	Desiccation	Many	Infiltration, lithology	Ready for field testing
Stabilizing Reagents	Gaseous Phosphate Injection	Pb, Cd, U, ⁹⁰ Sr	microbial activity, plume composition	Commercially available
Stabilizing Reagents	H ₂ S Injection	Cu, Ni, Cd, Hg, Pb	mineralogy	Field tested

Figure 5